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THE UTILIZATION OF MICROWAVE HEATING FOR THE FABRICATION OF SINTERED REACTION-BONDED SILICON NITRIDE

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ABSTRACT

The results of studies in which microwave heating was used to fabricate sintered reaction-bonded silicon nitride (SRBSN) are reviewed. These results are compared to parallel studies where conventional heating was used for the fabrication of these materials. Microwave fabrication of SRBSN involves a single heating cycle, whereas conventional processing requires two separate furnace runs and sample packaging steps. SRBSN containing high levels of sintering aids which were fabricated by microwave heating showed improved strength and toughness, as compared to those materials fabricated using a conventional resistance-heated furnace. An analysis of the microstructures of the microwave fabricated materials showed enhanced acicular grain growth as compared to conventionally heated material. Results are presented on studies involving the scale-up of the microwave fabrication process.

INTRODUCTION

Silicon nitride materials represents a class of materials having a wide range of compositions where silicon nitride (Si_3N_4) is the major phase. These materials are of interest in numerous applications for such diverse items as cutting tools, rotors and stator vanes for advanced gas turbines, valves and cam roller followers for gasoline and diesel engines, and radomes on missiles to name a few.[1] However, these materials tend to be very expensive and are not competitive with metal parts on a replacement basis.[2]

SRBSN has been identified as a cost-effective alternative to Si_3N_4 made from high cost raw materials. Silicon is economical compared to high purity Si_3N_4 powders (approximately 1/4 the cost). In addition, SRBSN materials are attractive in that they show improved control over the dimensional tolerances due to less shrinkage during sintering.[3,4]

In the last few years, microwave heating has been investigated extensively for the thermal processing of ceramics.[5-8] In addition, the microwave processing of Si_3N_4 has been the focus of considerable research.[9-11] The primary goal of this study is the investigation into the applicability of microwave heating for the fabrication of cost-effective SRBSN materials.

EXPERIMENTAL PROCEDURES

Three types of green preforms for the fabrication of SRBSN were used in this study: preforms fabricated by ORNL (ORNL TM-145), preforms fabricated by St. Gobain/Norton Industrial Ceramics (SG/NIC) as part of a cooperative research agreement, and bucket tappet preforms purchased from the Cremer Forschungsinstitut (CFI) of Germany. The details concerning the composition, size, shape, and preparation of these green preforms have been reported.[12,13,14]

Microwave processing of the preforms was conducted in a 500 L cylindrical microwave cavity with a 2.45 GHz power generator. These experiments involved the processing of from 2 to 40 samples, with masses ranging from 30 to 100 g each. The microwave nitridation and sintering of preforms was conducted in a single heating cycle, whereas the conventional processing of the preforms was carried out in a graphite-resistance heated furnace in a two-step process. The details concerning the conventional and microwave processes have been reported.[13,14]

Densities of sintered pieces were determined using the Archimedes method. Final microstructure of polished and plasma etched surfaces of sintered samples was examined with a Hitachi S-800 SEM. Selected samples of high density were machined into bend bar specimens with nominal dimensions of 3 mm x 4 mm x 50 mm, and the flexural strength testing was done in four point bending with inner and outer spans of 20 mm and 40 mm, respectively. Fracture toughness values were measured using Chantikul's method of four point bending of specimens indented with a Vickers indenter at a 10 kg load.[15]

RESULTS

Table 1 gives results including densities, flexural strengths, and fracture toughness values, characterizing ORNL TM 145 and SG/NIC SRBSN materials which were fabricated by either conventional or microwave heating. Two different trends were observed for the densities of the SRBSN produced using either microwave or conventional heating. For the SRBSN materials with high additive contents, the densities of the microwave processed materials were higher than the conventional samples . However, for the SRBSN containing low additive contents there was very little difference in the densities. This difference between the densities of the SRBSN matrials containing low and high additives which were processed by microwave heating is attributable to the microwave coupling effect inherent in the SRBSN. Pure Si_3N_4 is relatively transparent to microwaves, and microwave absorption during sintering is achieved through microwave coupling to sintering additives (e.g. Al_2O_3 and Y_2O_3) and nitridation enhancing additives (e.g. Fe_2O_3).[16] When the amount of additives is low, the SRBSN is difficult to heat, which translates into lower densities; and when the amount of sintering additives is high, higher densities are obtained. Also, Table 1 gives a comparison of the strength and toughness for the SRBSN materials produced by conventional and microwave processing. Similar trends to those observed with the densities exist for the strength and toughness values. Strength and toughness values for SRBSN materials containing high sintering aids and processed by microwave heating were higher than their conventional counterparts. Again, the reverse trend was true, in that the strength and toughness of the SRBSN materials with low sintering aids were comparable, irrespective of processing method.

Figure 1 shows SEM photomicrographs of polished and plasma etched surfaces of the sintered ORNL TM-145 SRBSN materials which were processed using conventional and microwave heating. The SRBSN material fabricated using microwave heating shows a much larger number of acicular grains than the SRBSN material fabricated with conventional heating. This phenomena may due to the preferential heating of the intergranular sintering aids which occurs during microwave processing of SRBSN.

Following the finding that microwave heating can be used to fabricate SRBSN materials with improved mechanical properties, work began on developing methods for the scale-up of this process. In all tests reported here, preforms for testing consisted of bucket tappets. Table 2 shows results obtained from three scale-up tests. These results show that for small numbers of samples, 8 to 9 bucket tappets, similar results were obtained for the conventional and microwave processes, however when the number of bucket tappets was increased to 21 in the microwave test, the average density decreased and the standard deviation of the densities increased. The reason for the decrease in the average density of the bucket tappets was found in an analysis of sample density versus sample location inside the microwave package. This analysis revealed that higher densities were obtained for bucket tappets in the center of the microwave package, and lower densities were obtained for samples at the periphery. It appears that sample orientation and proper insulation are critical for achieving uniform nitridation and densification of the preforms. Conventional scale-up processing runs with large numbers of samples have not been performed. Figure 2 shows a photo of bucket tappets nitrided and sintered using microwave heating in a scale-up run. Further tests runs are in progress to evaluate various microwave scale-up configurations.

Table 1. Summary of results on microwave and conventional fabrication of SRBSN.

Sample Number / Type	Sintering Aid Content	Processing Method	Sintered Density (g/cm^3)	Flexural Strength (MPa)	Fracture Toughness (K_{Ic}, $MPa\sqrt{m}$)
ORNL TM145	High[a]	Microwave	3.30	744	7.1±0.2
ORNL TM145	High[a]	Conventional	3.25	601	5.0±0.3
SG/NIC	High[b]	Microwave	3.31	876±33	7.3±0.1
SG/NIC	High[b]	Conventional	3.26	694±44	6.5±0.1
SG/NIC	Low[b]	Microwave	3.22	808±130	7.4±0.1
SG/NIC	Low[b]	Conventional	3.26	804±51	7.0±0.1

[a] 9 wt. % Y_2O_3 - 3 wt. % Al_2O_3 [b]proprietary composition

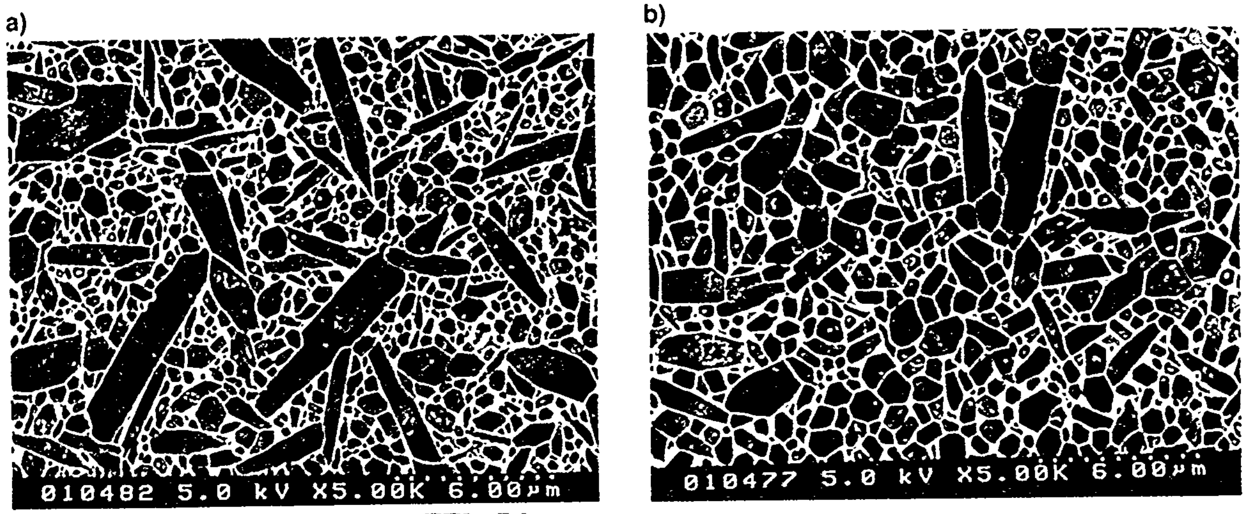

Figure 1. SEM photomicrographs of SRBSN materials fabricated using a) microwave heating and b) conventional heating.

Table 2. Results from three scale-up experiments

Processing Method	# Samples*	Avg Dens (g/cm^3)	St Dev Dens (g/cm^3)
Microwave	8	3.27	0.005
Microwave	21	3.23	0.022
Conventional	9	3.26	0.003

* bucket tappet samples from Cremer Forschungsinstitut, Germany

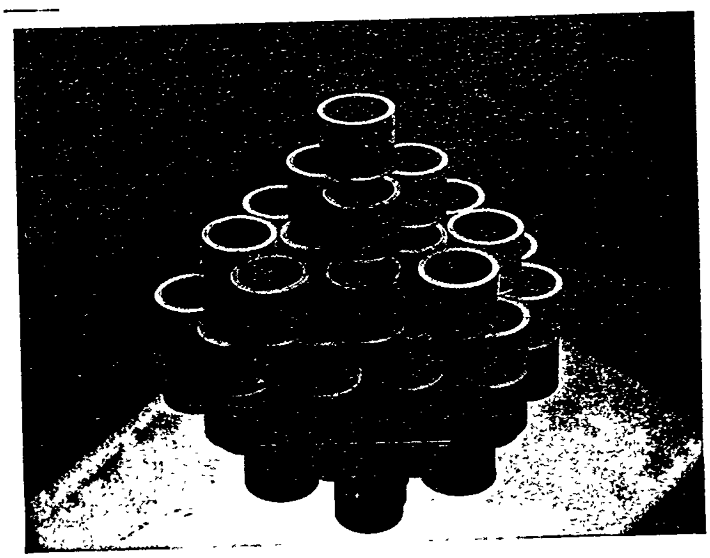

Figure 2. Bucket tappet samples nitrided and sintered using microwave heating in a scale-up test.

CONCLUSION

SRBSN is a cost-effective silicon nitride material. Raw materials costs are less than 1/4 those for high-purity silicon nitride materials, which improves the cost-competitiveness of these materials with metal parts. Conventional SRBSN requires long nitridation times and two-step firing. By using microwave heating, nitridation times are reduced and all firing is performed in a one-step continuous process. SRBSN materials with high levels of sintering aids which were fabricated by microwave heating show improved strength and toughness values, as compared to conventionally heated materials. Scale-up tests for the fabrication of complex-shaped SRBSN materials using microwave heating are in progress.

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